CANADIAN JOURNAL OF RESEARCH

VOLUME 22 MAY AND JULY, 1944 NUMBERS 3 AND 4

— SECTION A —

PHYSICAL SCIENCES

Contents

NATIONAL RESEARCH COUNCIL
OTTAWA, CANADA

CANADIAN JOURNAL OF RESEARCH

The Canadian Journal of Research is issued in six sections, as follows:

A. Physical Sciences
B. Chemical Sciences
C. Botanical Sciences
D. Zoological Sciences
E. Medical Sciences
F. Technology

For the present, each of these sections is to be issued six times annually, under separate cover, with separate pagination.

The *Canadian Journal of Research* is published by the National Research Council of Canada under authority of the Chairman of the Committee of the Privy Council on Scientific and Industrial Research. The *Canadian Journal of Research* is edited by a joint Editorial Board consisting of members of the National Research Council of Canada and the Royal Society of Canada.

Manuscripts should be addressed:

Editor-in-Chief,
Canadian Journal of Research,
National Research Council, Ottawa, Canada.

Canadian Journal of Research

Issued by THE NATIONAL RESEARCH COUNCIL OF CANADA

VOL. 22, SEC. A. MAY AND JULY, 1944 NUMBERS 3 AND 4

TRANSMISSION OF LIGHT BY WATER DROPS 1 TO 5 μ IN DIAMETER[1]

BY R. RUEDY[2]

Abstract

The size of the drops formed when water vapour condenses is related to the work performed against the surface tension. For the determination of the size by optical methods, Mie's solution of the electromagnetic equations for the propagation of light of wave-length λ in a medium containing small spherical drops of radius a indicates that as the ratio $\alpha = 2\pi a/\lambda$ increases from 0 to 20, the intensity of the light received in the prolongation of the incident beam passes alternately through maximum and minimum values. At distances from the drops greatly exceeding λ, the first maximum lies close to $\alpha = 2\pi$, the second is near $\alpha = 8.6$, and the subsequent peaks are less distinct and tend to repeat themselves at $\alpha = (m + 3/4)\pi$. As a result of these fluctuations the light seen through a cloud of particles with diameters greater than about 1μ is coloured. The theory accounts for the cycles in the changes of colour observed when the diameter increases, and enables a determination of the radius of growing drops. With increasing radius, the influence of the index of refraction m decreases; for $m = \infty$ the positions and values of the peaks differ only slightly from those obtained with water.

Introduction

Drops of water with a radius between 1 and 5 μ are coarser than the drops obtained in cloud chambers but finer than those in natural mist and fog. While drops of this intermediate size are of little practical interest because they are obtained only in the laboratory in a state sufficiently steady and uniform to allow detailed observation, they form nevertheless a necessary stage in the growth of drops and attract attention by their colours. They may, moreover, play a part at concentrations and at temperatures not usually encountered in the earth's atmosphere.

Since Hirn had verified experimentally, in 1862, that the sudden adiabatic expansion of dry saturated steam is accompanied by condensation, and Coulier and Aitken (9) had shown the importance of nuclei, the normal method of producing drops having a radius of a few microns is suddenly to change the volume of a given quantity of saturated and dust-laden air. With normal pressure and temperature initially, the changes in volume must be such that the pressure is reduced by at least 2 cm. of mercury and, if the

[1] *Manuscript received March 28, 1944.*

Contribution from the Research Plans and Publications Section, National Research Laboratories, Ottawa, Canada. Issued as N.R.C. No. 1211.

[2] *Research Investigator.*

The present article is an amplification of notes presented in 1931 at the Annual Meeting of the Royal Society of Canada.

adiabatic nature of the change is to be preserved, by not more than about 20 cm. of mercury. The resulting temperature drop, from $T_0°$ to $T°$ K, causes a cloud of small drops to form which may remain suspended for some time. Dust particles serve as centres upon which condensation begins. Their number varies from less than 1000 per cc., the concentration found in very pure air far away from inhabited districts, to as many as 50,000 per cc., a number readily obtained with air indoors.

The amount of vapour remaining in the air after an adiabatic expansion can be deduced from the equation for constant entropy (6, p. 729)

$$w \frac{L}{T} = w_0 \frac{L_0}{T_0} = Wc \ln \frac{T}{T_0}$$

where

w_0 is the mass of vapour per cubic centimetre before expansion,

w is the remaining mass of vapour,

W is the total quantity of liquid water and vapour per cubic centimetre,

c is the specific heat of water,

and

L is the latent heat of evaporation of water, equal to $594.9 - 0.51t$, with t in °C.

In cloud chambers, the lowest temperature T reached, after the expansion, is determined by the expansion of the much larger quantity of air mixed with vapour, and is deduced from the equation

$$\frac{T}{T_0} = \left(\frac{p}{p_0}\right)^{\frac{\kappa-1}{\kappa}} = \left(\frac{v_0}{v}\right)^{\kappa-1},$$

where $\kappa = 1.4$—unless the latent heat of the water condensed is capable of raising the temperature of the air.

If, for instance, the room temperature is about 17° C., a sudden reduction in the pressure from 75 cm. to 60 cm. of mercury produces a reduction in temperature from about 290° K to 272.7° K. The latent heat of evaporation of water is $594.9 - 0.51t$, so that $L = 586.3$ cal. and $L = 595.1$ cal. With these values the formula for w shows that $w = 0.955\ w_0$, or that 4.5% of the vapour is condensed. Since at 17° C. the amount of water vapour is at the most 14.5×10^{-6} gm. per cu. cm., the quantity condensed equals 535×10^{-6} gm. per gm. air. In its condensation this quantity yields 0.3 cal. of latent heat, and increases the temperature of the air ($C_p = 0.237$ cal. per gm. per deg. C.) by 1.3° so that the lowering amounts to 16.0° instead of 17.3° C. When the amount of condensed water is recalculated for the corrected temperature it is found that $w = 0.957\ w_0$, in other words that only 4.3% of the moisture is condensed by a single expansion despite the fact that as a result of the rapid decrease in vapour pressure with falling temperature the water vapour becomes markedly supersaturated during the expansion. If the density of saturated water vapour at $T°$ K is ρ_0, and at $T°$ it is ρ ($\rho = 0.485$ gm. per cc. at 0° C.), and the supersaturation S is defined

as the ratio of the actual density of the vapour to the density of the saturated vapour at the same temperature T, then

$$S = \frac{\rho_0}{\rho}\frac{v_0}{v} = \frac{\rho_0}{\rho}\left(\frac{T_0}{T}\right)^{\frac{1}{\kappa-1}}.$$

For an expansion reducing the temperature from 290 to 273° K, S equals 3.5 and higher values are obtained for greater ratios of expansion.

This lag in the condensation of water vapour is only in part accounted for by the larger vapour pressure of small drops and their tendency to evaporate as quickly as they are formed. The main reason is that the formation of a drop requires mechanical work of an amount greatly exceeding the work that can be performed by molecules or clusters of molecules at normal pressures and temperatures. The following calculation of the vapour pressure of small drops yields not only the correct pressure according to Lord Kelvin (6), but also the amount of work to be carried out during the formation of drops.

If at the temperature T a drop of radius r has a vapour pressure p_r and a larger drop of radius r_1, at the same temperature, is in equilibrium with vapour at the pressure p_1, the work required for the compression of a small quantity dw of vapour from the pressure p_r to the smaller pressure p_1 is equal to $dw\,RT \ln (p_r/p_1)/M$, where M is the molecular weight of the substance and R is the gas constant (8.314×10^7 ergs per deg.). It may be imagined that this quantity dw consists of molecules that have escaped from the smaller drop and that after the compression condense at the same temperature upon the larger drop. Such a transfer involves a decrease $4\pi r^2\,dr$ in the volume of the smaller drop, and an increase $4\pi r_1^2\,dr_1$ in the volume of the larger drop. For this change $dw = 4\rho\pi r^2 dr$ or $dr = dw/4\pi r^2\rho$. The surface of the smaller drop loses an area equal to $8\pi r\,dr$; the larger drop gains $8\pi r_1\,dr_1$ in area. The reduction in area produces work equal to $2\sigma dw/r\rho$ erg, whilst the increase in the surface of the larger drop absorbs the amount $2\sigma dw/r_1\rho$ erg, where σ is the surface tension in dynes per cm. The difference must be balanced by the work done during compression, so that Lord Kelvin's formula follows:

$$\ln\frac{p_r}{p_1} = \frac{2\sigma}{RT}\frac{M}{\rho}\left(\frac{1}{r} - \frac{1}{r_1}\right).$$

When $r = \infty$, that is, when a flat surface of the liquid is considered, the equation gives the ratio between the vapour pressure of the drop of radius r and the vapour pressure over the liquid when present in bulk.

The work required for producing from the supersaturated vapour a drop that has a radius r and a corresponding vapour pressure p_r is deduced by considering that in the experiment conducted the mass of water lost by the small drop is equal to the mass acquired by the large drop; hence

$$dw = 4\rho\pi r^2\,dr = 4\rho\pi r_1^2\,dr_1$$

or

$$dr_1 = \frac{r^2}{r_1^2}\,dr$$

and the work required for the small change dr in radius is

$$dA = \sigma\left(8\pi r - 8\pi\frac{r^2}{r_1}\right)dr.$$

When this quantity is integrated from the value r corresponding to the smallest drop, practically zero, to a value r, the result

$$A = \left(12\pi\frac{r^2}{3} - \frac{8\pi r^3}{3r_1}\right)\sigma$$

gives the work that has to be done against the surface tension when a drop of radius r is to be formed. The fraction of the molecules that possess kinetic

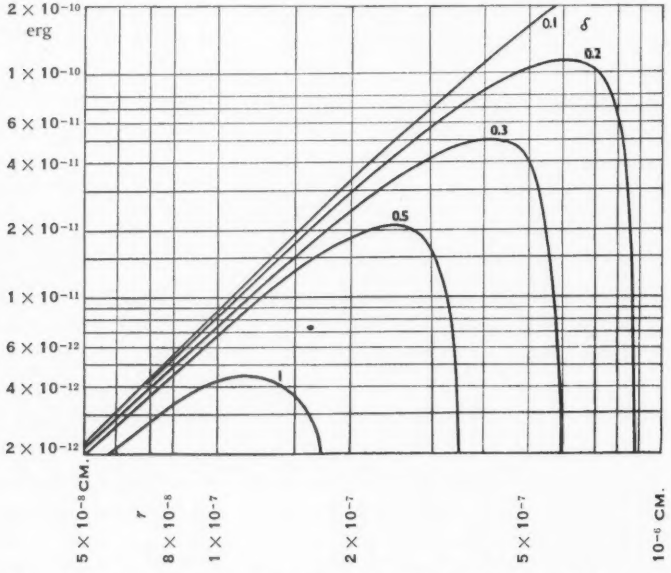

FIG. 1. *Work to be performed against the surface tension in the formation of drops of radius r at various degrees of supersaturation S or* $\ln S = \delta$ *at 0° C.*

energy equal to A ergs is $\epsilon^{-A/kT}$. Since according to Lord Kelvin's equation for the vapour pressure of small drops that are in complete equilibrium, the radius obeys the relation

$$r = \frac{2\sigma M}{\rho RT}\frac{1}{\delta},$$

where $\delta = \ln p_r/p_\infty$, the equation for the work required may be written

$$A = 4\pi r^2\left(\sigma - \frac{r}{3}\frac{\rho RT}{M}\delta\right).$$

For water drops of radius r the work of formation near room temperature ($T = 300°$ K) is

$$A = 4\pi r^2(71.7 - 46.19\, r\, 10^7\delta).$$

A study of this equation represented in Fig. 1 for $T = 273.2°$ K, reveals that at ordinary temperatures the molecules have insufficient energy to enable the formation of drops of other than the most minute size. The ratio A/kT of the mechanical work for the formation of a drop of radius r, to the average kinetic energy of one molecule per degree of freedom, decreases to small values only when the temperature is increased (Table I) or when the mixture is

TABLE I

VAPOUR PRESSURE OF SMALL DROPS OF WATER 1μ AND LESS IN DIAMETER AT 0° C. (σ = 75.6 DYNES PER CM.) AND AT 75° C. (σ = 63.5 DYNES PER CM.)

Radius a, cm.	0° C.		75° C.	
	$\frac{p_r}{p_\infty}$	$\frac{A_0}{kt}$	$\frac{p_r}{p_\infty}$	$\frac{A_0}{kt}$
10^{-4}	1.001	8.4×10^7	1.001	5.5×10^7
5×10^{-5}	1.002	2.1×10^7	1.003	1.4×10^7
10^{-5}	1.012	8.4×10^5	1.012	5.5×10^5
5×10^{-6}	1.025	2.1×10^5	1.025	1.4×10^5
10^{-6}	1.128	8.4×10^3	1.131	5.5×10^3
5×10^{-7}	1.273	2100	1.280	1.4×10
2×10^{-7}	1.827	336	1.854	221
10^{-7}	3.34	84	3.437	55.4
5×10^{-8}	11.1	33.6	11.8	13.8
2.5×10^{-8}	124	5.25	140	3.5
		$\epsilon^{-5.25} = 0.005$		$\epsilon^{-3.5} = 0.030$

(The fraction σ/ρ is assumed to depend only on T.)

flowing at a very high speed. It is therefore natural that condensation occurs but sparingly and slowly even in air that is highly supersaturated. Thus while Lord Kelvin's vapour pressure equation indicates that a supersaturated vapour of given temperature is in equilibrium with drops of only one size, the work required for the formation of particles of finite size prevents, in general, the establishment of this equilibrium. Were it not for this lag, the radius of small drops could be deduced directly from the vapour pressure equation. Near the boiling point at atmospheric or higher pressure this method seems to give results that are at least of the correct order of magnitude (10). It may be recalled, in this connection, that the increase in vapour pressure with decreasing size of the drops is the same as the increase observed over an extended flat water surface when the external pressure is increased by an amount equal to the internal pressure, σ/r, created in the drop by the surface tension.

Determination of the Size of the Drops

On account of the small amount of water condensed in a single expansion, several repetitions of the experiment are possible after the drops have been allowed to settle and the mixture of air and vapour has been brought back to

its original volume. A second like expansion results in the formation of a similar cloud; after the experiment has been repeated a number of times, the cloud becomes thinner and the drops are less numerous but, because about the same quantity of vapour condenses each time, they are correspondingly larger. At a certain stage the expansion produces a rain of quickly settling large drops, and in the end the expansion ratios, and consequently the degree of supersaturation, have to be raised before condensation recurs. In Wilson's work with cloud chambers the expansion ratio v/v_0 had to be increased from 1.25 before the rain stage was reached, to 1.4 after the precipitation of large drops (9). The water is then obtained in the form of a persistent cloud of small drops. The number of droplets is larger the larger the expansion ratio v/v_0.

The general explanation of the cloud chamber experiments is well known. The clouds first formed are produced by condensation on the dust particles always present in the atmosphere; the number of nuclei decreases at each expansion because they settle to the bottom with their envelope of water, and, in the end, condensation is possible only on clusters of water molecules forming in the water vapour, or on the smallest specks of dust that remain suspended in the air. The available kinetic energy of the molecules allows only the formation of small drops. With the tiniest drops, colours begin to border the image of a source viewed through the mass of condensing particles, and when according to rough estimates the diameter of the particles approaches the wavelength of light, the centre of the strongly blurred image of the source, which was white in the presence of the larger particles, becomes distinctly coloured.

Whether dust particles facilitate condensation merely by eliminating the part of the work to be carried out against the surface forces in creating a drop of the size of the dust particle, or whether the heat of wetting assists condensation, are questions that remain to be decided. The difference D in the work of producing a drop of large radius r_0 and that of producing a drop of radius r_1 is

$$D = 4\pi\sigma\,(r_0^2 - r_1^2) - 4\frac{\pi}{3}\frac{RT}{M}\,\rho\delta\,(r_0^3 - r_s^3)$$

$$= (r_0 - r_1)\left\{4\pi\sigma(r_0 + r_1) - 4\frac{\pi}{3}\frac{RT}{M}\,\rho\delta(r_0^2 + r_0r_1 + r_1^2)\right\}$$

or

$$D \doteq 4\pi\sigma(r_0^2 - r_1^2) = 4\pi\sigma r_0^2\left(1 - \frac{r_1^2}{r_0^2}\right).$$

In other words, the reduction in the work obtained by the substitution of a ready made particle in place of a liquid drop of the same size is appreciable only if the radius of the solid nucleus is almost as large as that of the equilibrium drop. The slight gain obtained when $r_1 < r_0/2$ suggests that the forces of adhesion intervene in the formation of drops upon dust nuclei.

The colours produced by these small drops were analysed in detail on several occasions (2, 3, 7). The adiabatic expansion is initiated either by the

sudden rotation of a stopcock connecting a container of several litres capacity in which the saturated air is kept, with a partially evacuated large flask, or by the rapid displacement of a tightly fitting small piston, lubricated with water. The air used in the experiment need not be filtered. If it happens to contain too many dust particles, a few strong expansions reduce their number and increase the amount of water condensed upon each nucleus (3). Fewer and therefore larger drops are formed at each step, and the whole range of diameters, from a small fraction of a micron after the first experiments to a few microns, the rate of increase being about one-tenth micron at each expansion, is obtained in this way and is available for study during about one minute. The drops are in a labile equilibrium, because the slightest decrease in radius in one particle causes increased evaporation and leads to the disappearance of the drop, whilst the larger drops tend to grow at the expense of the vapour lost by the small drops. It is reported, however, that if instead of reducing the number of dust particles by deposition at the end of each expansion, a method likely to decrease not only the number but also the size of the particles, a measured quantity of dust-laden air from a large storage vessel is introduced before every test so that the size of the dust nuclei remains constant, the mist formed is quite stable for at least an hour (4). Kiessling, one of the first investigators to suggest that a relation exists between the diameter and the colour in which the light source appears, found that as larger drops are produced, a source of white light transmits first pale lilac, then blue-lilac, bright light blue, bluish green, emerald green, yellow-green, light orange, dark orange, scarlet, crimson, stone gray, olive green, yellow-green, bronze, orange, etc. (2). In addition, the central coloured image of the source seen by the eye appears to be surrounded by coloured rings. The tint of the rings varies with increasing diameter of the particles in a seemingly irregular manner. Detailed observations give the following succession of colours (3).

Central Field	*Outer Rings*
First Cycle	
Light violet	Green
Blue violet	Yellow green
Pale blue	Green, pink
Blue (greenish)	Dark blue, red
Green blue	Light violet (reddish)
Green	Violet blue, light green, pink
Yellow green	Blue, pink
Yellow	Green, violet
Orange	Green, violet
Red	Green, violet
Red	Pale green (yellowish) pink
Purple	Pale green, pink
Light purple	Green, reddish, violet
Second Cycle	
Violet	Green, red, green
Violet	Yellow, red, dark blue
Grey-blue	Green, dark violet, yellow
Blue-green	Yellow, violet, green
Bluish green	Yellow, green, pink

Central Field	*Outer Rings*
Second Cycle—*Concluded*	
Emerald	(Pure green), dark blue, red
Olive green	Dark blue, red
Greenish yellow	Blue, green, red, violet
Greenish yellow	Red, blue, green
Yellow	Red, green, violet
Yellow	Black (pale blue, green, red, blue) red
Light orange	Red, green, violet
Light purple	Yellow green, pink

The central field of the first cycle is wider than that obtained in the second cycle, and its colours are saturated but not very bright. In the second cycle the intensity of the light is much greater; the diameter of the field and of the rings is reduced so that a most striking assortment of colours is seen. The third cycle is a repetition of the first two with still further reduced areas. The colour of the central field is paling, and the rings are so closely packed that it is difficult to distinguish the separate tints.

In Wilson's first cloud chamber experiments the same cycles of colour were observed, but in the reverse order (9). He increased the rate of expansion at each step, and apparently as the supersaturation was increased, a larger number of clusters of water molecules came into play so that each received a smaller share of the water that condensed, and smaller drops resulted with increased expansion. An observer using this method and looking through the cloud chamber slightly to one side of the source of light sees the central part of the illuminated field coloured whenever the ratio v/v_0 equals 1.4. After the ratio has reached 1.41 the field is bright green; at 1.416 it is blue green; at 1.418, brilliant blue, then violet, and at $v/v_0 = 1.420$, reddish-purple or mauve, this being the so-called sensitive tint. Increases in steps of 0.005 in the expansion ratio cause the colour to change from the sensitive tint to red, then to reddish-yellow and to yellowish white. The supersaturation S required to produce the first trace of colour is 7.9; that for producing the sensitive tint is equal to 9.9.

Since the calculation of the intensities of the light behind spherical obstacles was judged to be too difficult if the rigorous theory developed by Mie was used for the purpose, several attempts were made hitherto to derive a relation between colour and drop diameter by approximate methods. In one of the most detailed studies (3) the light received at any one point remote from the obstacle was considered to be the result of three contributions, namely, first the plane waves of light received by diffraction around an opaque disk occupying the cross-section of the sphere and placed normal to the incident ray; second, light transmitted through the spherical particle after two refractions, the intensities being reduced at each refraction according to Fresnel's laws as well as by the light spreading out in cones that have their summits at the focus of the drop considered as a spherical lens; third, light reflected at the surface of the drop, in particular from the portion receiving it at glancing incidence. The contribution furnished by diffraction comes mainly from a narrow ring surrounding the sphere where the disk intersects the particle.

Refraction adds to the intensity in the prolongation of the incident ray; reflection represents nearly parallel rays that come mainly from a narrow rim surrounded by the ring responsible for diffraction. Depending on the phase differences at a given wave-length, the reflected and the refracted rays may assist or cancel each other. Their contribution depends on a factor, $(\pi a^2 \sin^2 \gamma)^{-1}$, which is inversely proportional to the area of the cross-section and tends toward zero when the radius of the drop attains large values.

There is no doubt that by the inclusion of refraction and reflection a more accurate representation of the actual conditions about the sphere is obtained than when the drop is considered merely as an opaque obstacle placed in the path of the incident light. There remains no doubt, on the other hand, that this approximate theory fails when the diameter is smaller than 1μ, and since there seems to be no way of defining its range of validity, the one reliable method appears to be the calculation according to the exact theory and the comparison of the results with the simplified formula (7).

For the calculations, the expressions for the amplitudes of the electric and magnetic components of the light wave are preferably written as

$$a_\nu = \frac{(-1)^\nu (2\nu+1)}{-i-(-1)^\nu \dfrac{J_{-\nu-\frac{1}{2}}(\alpha)}{J_{\nu+\frac{1}{2}}(\alpha)} \dfrac{\dfrac{J_{-\nu+\frac{1}{2}}(\alpha)}{J_{-\nu-\frac{1}{2}}(\alpha)} + \dfrac{1}{m}\dfrac{J_{\nu-\frac{1}{2}}(\beta)}{J_{\nu+\frac{1}{2}}(\beta)} + \dfrac{\nu}{\alpha}\left(1-\dfrac{1}{m^2}\right)}{\dfrac{J_{\nu-\frac{1}{2}}(\alpha)}{J_{\nu+\frac{1}{2}}(\alpha)} - \dfrac{1}{m}\dfrac{J_{\nu-\frac{1}{2}}(\beta)}{J_{\nu+\frac{1}{2}}(\beta)} - \dfrac{\nu}{\alpha}\left(1-\dfrac{1}{m^2}\right)}}$$

$$p_\nu = \frac{(-1)^{\nu+1}(2\nu+1)}{-i-(-1)^\nu \dfrac{J_{-\nu-\frac{1}{2}}(\alpha)}{J_{\nu+\frac{1}{2}}(\alpha)} \dfrac{\dfrac{J_{-\nu+\frac{1}{2}}(\alpha)}{J_{-\nu-\frac{1}{2}}(\alpha)} + m\dfrac{J_{\nu-\frac{1}{2}}(\beta)}{J_{\nu+\frac{1}{2}}(\beta)}}{\dfrac{J_{\nu-\frac{1}{2}}(\alpha)}{J_{\nu+\frac{1}{2}}(\alpha)} - m\dfrac{J_{\nu-\frac{1}{2}}(\beta)}{J_{\nu+\frac{1}{2}}(\beta)}}}$$

where

$\alpha = 2\pi a/\lambda$

$\beta = m\alpha$

J is the symbol for the Bessel function of the first kind.

The following relations between Bessel functions are useful for checking the values of $J_{-\nu-\frac{1}{2}}/J_{\nu+\frac{1}{2}}$, $J_{-\nu+\frac{1}{2}}/J_{-\nu-\frac{1}{2}}$, and $J_{\nu-\frac{1}{2}}/J_{\nu+\frac{1}{2}}$.

$$\frac{J_{-\nu-\frac{1}{2}}}{J_{\nu+\frac{1}{2}}} = \frac{-\dfrac{J_{-\nu+\frac{1}{2}}}{J_{\nu-\frac{1}{2}}}}{1-\dfrac{\alpha}{2\nu-1}\dfrac{J_{\nu-3/2}}{J_{\nu-\frac{1}{2}}}} + \frac{\dfrac{J_{-\nu+3/2}}{J_{\nu-3/2}}}{1-\dfrac{2\nu-1}{\alpha}\dfrac{J_{\nu-\frac{1}{2}}}{J_{\nu-3/2}}}$$

$$\frac{J_{\nu+\frac{1}{2}}}{J_{\nu+3/2}} = \frac{1}{-\dfrac{J_{\nu-\frac{1}{2}}}{J_{\nu+\frac{1}{2}}} + \dfrac{2\nu+1}{\alpha}}$$

$$\frac{J_{-\nu-\frac{1}{2}}}{J_{-\nu-3/2}} = \frac{-1}{\dfrac{J_{-\nu+\frac{1}{2}}}{J_{-\nu-\frac{1}{2}}} - \dfrac{2\nu+1}{\alpha}}.$$

The ratio between the intensity of the light in the prolongation of the incident beam, and the intensity of the incident light, at points not close to the drop of water, is given by the formula

$$I_1 = \frac{\lambda^2}{4\pi^2 r^2} \left| \Sigma\, (-1)^{\nu+1}(a_\nu - p_\nu) \right|^2 .$$

In the derivation of this expression it is assumed that the incident light is a plane wave and completely polarized, the electrical vibrations being parallel to the z-axis, while scattered light is observed in the plane x, y at right angles to z. The same formula holds when the electrical vibrations are confined to the plane of observation x, y and are at right angles to the direction x along which the plane wave advances.

In ordinary, unpolarized light, the intensity of the component vibrating at right angles to the plane of observation is equal to that of the component parallel to this plane. If the intensity is equal to unity for each of the two components, half the sum $(I_1 + I_{11})$ usually represented in tables or graphs is a correct measure of the intensity of the light scattered in the direction $\gamma = 180°$, that is, from the source to the particle.

The values of a_ν and p_ν increase from a small value for small values of ν to a maximum when ν is approximately equal to α, and decrease rapidly when ν exceeds α by one or two units. The results of the calculations of I are represented in Fig. 2 together with a few values previously obtained (1, 4). The factor $\lambda^2/4\pi^2r^2$ has been taken equal to unity; in practice it has much smaller values since, if the theory is to hold, the observations have to be made at distances r large compared with the wave-length.

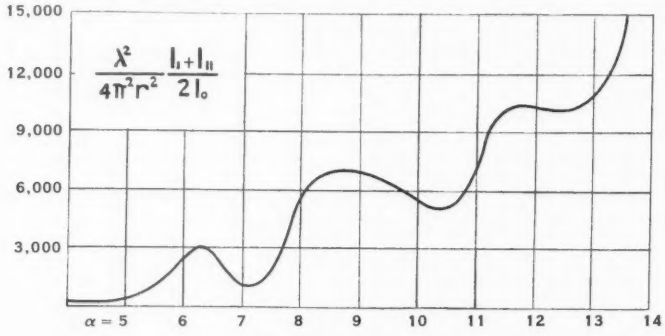

FIG. 2. *Fraction $I/2I_0$ of light transmitted by a water drop as a function of the ratio $2\pi a/\lambda = \alpha$. The factor $\lambda^2/4\pi^2r^2$ has been put equal to unity, $I_0 = I_1 + I_{11}$.*

A glance at the curve illustrating the change in I as a function of α at once shows a first maximum near $\alpha = 2\pi$ and a second pronounced maximum near $\alpha = 8.5$. Less distinct peaks lie near $\alpha = 12$ and $\alpha = 15$. The first peak is such that when the value of α is equal to 6.28 for violet or blue light, this colour will be predominant in the central field when the eye looks through

the cloud of drops towards the source of light. As the diameter of the particles increases, α will attain the value 6.3 for wave-lengths longer than those in the violet or in the blue, and the colour of the central field will appear in turn green, orange, and red. The same cycle is repeated when the diameter has increased to a value for which α equals about 8.5 for violet light, except that the light transmitted has much greater intensity.

Application

In order to verify how accurately the theory accounts for the observed colour changes it is necessary to remember that the computed intensity for a given distance r is proportional to the square of the wave-length. For particles 1μ in diameter, for instance, the values of α are 5 at $\lambda = 0.630\ \mu$, 5.95 at $\lambda = 0.528\ \mu$, and 6.9 at $\lambda = 0.457\ \mu$. After multiplication with λ^2, the intensities at the three wave-lengths are about as 158 : 600 : 252. According to Maxwell's colour equation, white can be considered as a mixture of the colours corresponding to the three wave-lengths mentioned, providing that the intensities in the red, green, and violet are to one another as 240 : 383 : 377. Subtraction of an amount of white (67%) in which the intensities at $0.630\ \mu$, $0.528\ \mu$, and 0.457μ are in the correct ratio, namely, as 158 : 256 : 252, leaves green and blue in the transmitted light, so that according to Maxwell's colour triangle (5, p. 51) the central portion of the blurred image of a white source seen through the cloud of drops is coloured a light green blue. For slightly larger particles, 1.1μ in diameter, the subtraction of white light (53%) leaves coloured light consisting of 33% red and 67% green (yellow green).

Between 1.10 and 1.15μ diameter, the colours turn in rapid succession from yellow (at 1.12μ) through orange and red to purple (1.125μ). For 1.3μ the colour is reddish violet with 43% white; it then changes through violet to blue (about 1.35μ) back to greenish blue. For still larger drops a second cycle of colour changes begins, characterized by weakness in the red. The second green is pronounced when the diameter $2a$ equals 1.5μ. Through this range the colours of the central field are about the same as those of a thin parallel plate of thickness $2a(m-1)$ as was presumed many years ago by Barus (3).

Influence of the Index of Refraction

When the index of refraction passes from 4/3 to very large values, the formulae for the components a_ν and p_ν reduce to the following expressions

$$a_\nu = \frac{(-1)^\nu(2\nu+1)}{-i-(-1)^\nu \dfrac{J_{-\nu-\frac{1}{2}}(\alpha)}{J_{\nu+\frac{1}{2}}(\alpha)} \dfrac{\dfrac{J_{-\nu+\frac{1}{2}}(\alpha)}{J_{-\nu-\frac{1}{2}}(\alpha)} + \dfrac{\nu}{\alpha}}{\dfrac{J_{\nu-\frac{1}{2}}(\alpha)}{J_{\nu+\frac{1}{2}}(\alpha)} - \dfrac{\nu}{\alpha}}}$$

$$p_\nu = \frac{(-1)^{\nu+1}(2\nu+1)}{-i+(-1)^\nu \dfrac{J_{-\nu-\frac{1}{2}}(\alpha)}{J_{\nu+\frac{1}{2}}(\alpha)}}.$$

On carrying out the summation in order to arrive at the total intensity $I = I_1 + I_{11}$ in the prolongation of the incident ray, it is found that in contrast with the results obtained for smaller particles (Fig. 3), the index of refraction changes the intensities but slightly for particles with a diameter exceeding 1μ. For $\alpha = 5$, the intensity in the direction from the source through the particle to the observer decreases by 11% when the index of refraction increases from 4/3 to ∞, but for $\alpha = 10$, the reduction amounts to 4%. For $\alpha = 2\pi$, the difference is larger, namely, 40%; for $\alpha = 4\frac{1}{2}\pi$ it has decreased to 10%. For $\alpha > 20$ the index of refraction may be considered as having a practically negligible effect.

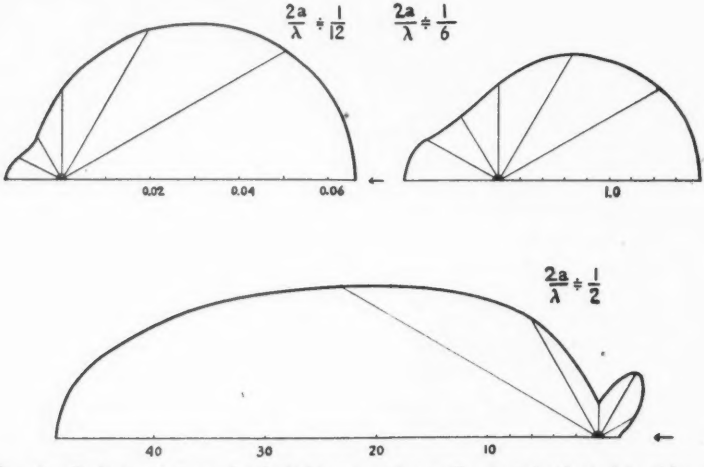

FIG. 3. *Radiation patterns for the light sent out by an illuminated spherical particle with a very large index of refraction, at various ratios $2a/\lambda$. The light source is at the right.*

The greater strength in the intensity of the light that a small particle sends back to the source (Fig. 3, top row) is not a feature peculiar to materials with an index of refraction tending to infinity. This same difference is found for all substances, as might be expected from the mere consideration that one side of the particle is directly illuminated while the other receives light only by diffraction. But the difference is most often quite negligible; the formulae for small values of α

$$a_1 = 2\alpha^3 \frac{m^2 - 1}{m^2 + 2}$$

$$p_1 = -\frac{\alpha^5}{15}(m^2 - 1)$$

$$a_2 = -\frac{\alpha^5}{6}\frac{m^2 - 1}{m^2 + 3/2}$$

show indeed that

$$a_1 \mp p_1 = \alpha^3 \frac{m^2 - 1}{m^2 + 2}\left(1 \pm \alpha^2 \frac{m^2 + 2}{30}\right).$$

The difference between $(a_1 + p_1)/2$ and $(a_1 - p_1)/2$ on which the intensities depend, becomes appreciable for $\alpha = 0.1$, for instance, when $(m^2 + 2)/30$ is about equal to 10, or m larger than 5. For larger values of α the component a_2 also must be taken into consideration.

The formulae for a_2 and p_2 derived for large values of the index of refraction m hold also when m is finite but the index of absorption is large and the particle is therefore completely opaque.

Moreover, whatever the values of the optical constants, provided only that α exceeds a certain value, there is complete agreement, at least for the directions to and from the source, between the values obtained for the intensities of the scattered light when the solutions of the electromagnetic theory of propagation of plane waves are used, or when the simpler expressions given by the theory of diffraction of light by opaque circular disks are taken. Indeed when α is large

$$a_\nu = \frac{2\nu + 1}{m + 1} i^\nu \left(\cos\alpha \sin\left(\alpha - \nu\frac{\pi}{2}\right) - m \sin\alpha \cos\left(\alpha - \nu\frac{\pi}{2}\right)\right)$$
$$+ \frac{2\nu + 1}{m + 1} i^{\nu+1}\left(m \cos\alpha \cos\left(\alpha - \nu\frac{\pi}{2}\right) + \sin\alpha \sin\left(\alpha - \nu\frac{\pi}{2}\right)\right)$$
$$-p_\nu = \frac{2\nu + 1}{m + 1} i^\nu \left(m \cos\alpha \sin\left(\alpha - \nu\frac{\pi}{2}\right) - \sin\alpha \cos\left(\alpha - \nu\frac{\pi}{2}\right)\right)$$
$$+ \frac{2\nu + 1}{m + 1} i^{\nu+1}\left(\cos\alpha \cos\left(\alpha - \nu\frac{\pi}{2}\right) + m \sin\alpha \sin\left(\alpha - \nu\frac{\pi}{2}\right)\right)$$

According to these expressions the real part in the sum of all the differences $(a_\nu - p_\nu)$ vanishes, while the imaginary part is equal to the sum of all the odd numbers up to $(2\nu + 1)$ and therefore equal to $(\nu + 1)^2$ or, since about $\nu = \alpha$ terms must be added, approximately equal to α^2. Hence

$$\frac{I_1 + I_{11}}{2} \approx \frac{\lambda^2}{4\pi^2 r^2}\frac{\alpha^4}{4} = \frac{a^2\alpha^2}{4\pi^2 r^2}.$$

According to the theory of diffraction of light by opaque disks having a radius a, the intensity of the light received in the direction forming the angle γ with the incident beam is

$$I = \frac{4\pi^2 a^4}{\lambda^2 r^2} \sin^4\gamma/2 \frac{J_1^2(\alpha \sin\gamma)}{(\alpha \sin\gamma)^2} I_0,$$

where I_0 is the intensity of illumination. In the direction from the source to the particle and to the observer ($\gamma = 180°$)

$$\frac{I}{I_0} = \frac{a^2\alpha^2}{4\pi^2 r^2}.$$

Both theories indicate therefore that for a given wave-length, the intensity along the extension of the incident beam increases with the fourth power of α, but as shown by the diffraction formula the intensity decreases rapidly to zero when γ decreases, and this reduction is faster for larger values of α.

High intensities are found only between 180° and a slightly smaller angle determined by the relation

$$\sin \gamma \doteqdot \gamma = \frac{1.22\pi}{\alpha} = 0.61 \frac{\lambda}{a}.$$

Apart from this reservation, drops of water of the size found in natural fogs behave like opaque particles; this result is in agreement with the view adopted in meteorology.

The Coefficient of Absorption

If there is one particle in unit volume, the coefficient of absorption k, which determines the ratio between the intensities of the transmitted light and the incident radiation according to the formula $I = \epsilon^{-kr} I_0$, at the end of a path of length r in air, is given by

$$k = \text{imaginary part of } 2\pi a^2 \sum_1^\infty \frac{a_\nu - p_\nu}{\alpha^2}.$$

The calculations give $k = 1.95$ for $\alpha = 2$, $k = 0.8$ for $\alpha = 11$, and $k = 1.5$ for $\alpha = 4\frac{1}{2}\pi$. The loss along the path of the light is caused almost exclusively by scattering. However since, with particles of this size, a fraction of the scattered light is radiated along the extension of the original direction of the light and restored to the emerging beam, a correction must be applied to the computed value of k. It is obtained by taking the average of the scattered intensity for the directions along which the light is received.

References

1. BLUMER, H. Z. Physik, 38 : 304-328, 920-947. 1926.
2. KIESSLING, J. Meteor. Z. 1 : 117-126. 1884.
3. MECKE, R. Ann. Physik, 61 : 471-500. 1920; 62 : 623-648. 1920.
4. PARANJPE, G. R., NAIK, Y. G., and VAIDYA, P. B. Proc. Indian Acad. Sci. A, 9 : 333-351, 352-364. 1939.
5. PEDDIE, W. Colour vision. E. Arnold & Co., London. 1922.
6. PRESTON, T. Theory of heat. 4th ed., ed. by J. R. Cotter. Macmillan and Co., Ltd., London. 1929.
7. RUEDY, R. Can. J. Research, A, 21 : 99-109. 1943.
8. SANDERSON, J. A. J. Optical Soc. Am. 30 : 405-409. 1940.
9. WILSON, C. T. R. Trans. Roy. Soc. (London) A, 189 : 265-307. 1897.
10. YELLOT, J. I. Trans. Am. Soc. Mech. Engrs. 56 : 411-426.; discussion, 427-430. 1934.

CANADIAN JOURNAL OF RESEARCH

Notes on the Preparation of Copy

General:—Manuscripts should be typewritten, double spaced, and the **original and at least one extra copy** submitted. Style, arrangement, spelling, and abbreviations should conform to the usage of this Journal. Names of all simple compounds, rather than their formulae, should be used in the text. Greek letters or unusual signs should be written plainly or explained by marginal notes. Superscripts and subscripts must be legible and carefully placed. Manuscripts should be carefully checked before being submitted, to reduce the need for changes after the type has been set. **All pages, whether text, figures, or tables, should be numbered.**

Abstract:—**An abstract of not more than about 200 words, indicating the scope of the work and the principal findings, is required.**

Illustrations

(i) *Line Drawings:*—Drawings should be carefully made with India ink on white drawing paper, blue tracing linen, or co-ordinate paper ruled in **blue** only. Paper ruled in green, yellow, or red should not be used. The principal co-ordinate lines should be ruled in India ink and all lines should be of sufficient thickness to reproduce well. Lettering and numerals should be of such size that they will not be less than one millimetre in height when reproduced in a cut three inches wide. If means for neat lettering are not available, lettering should be indicated in pencil only. All experimental points should be carefully drawn with instruments. Illustrations need not be more than two or three times the size of the desired reproduction, but the ratio of height to width should conform with that of the type page. **The original drawings and one set of small but clear photographic copies are to be submitted.**

(ii) *Photographs:*—Prints should be made on glossy paper, with strong contrasts; they should be trimmed to remove all extraneous material so that essential features only are shown. Photographs should be submitted **in duplicate;** if they are to be reproduced in groups, one set should be so arranged and mounted on cardboard with rubber cement; the duplicate set should be unmounted.

(iii) *General:*—**The author's name, title of paper, and figure number should be written on the back of each illustration.** Captions should not be written on the illustrations, but typed on a separate page of the manuscript. All figures (including each figure of the plates) should be numbered consecutively from 1 up (arabic numerals). **Reference to each figure should be made in the text.**

Tables:—Titles should be given for all tables, which should be numbered in Roman numerals. Column heads should be brief and textual matter in tables confined to a minimum. **Reference to each table should be made in the text.**

References should be listed alphabetically by authors' names, numbered in that order, and placed at the end of the paper. The form of literature citation should be that used in this Journal and titles of papers should not be given. All citations should be checked with the original articles. Each citation should be referred to in the text by means of the key number.

The *Canadian Journal of Research* conforms in general with the practice outlined in the *Canadian Government Editorial Style Manual*, published by the Department of Public Printing and Stationery, Ottawa.

Reprints

Fifty reprints of each paper are supplied free. Additional reprints, if required, will be supplied according to a prescribed schedule of charges.